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# Recovery of acidified Finnish lakes: trends, patterns and dependence of catchment characteristics

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## Abstract

The regional-scale trends (south, central and north Finland) in key acidification parameters over the period 1990–2003 were studied in lakes used for monitoring of acidification (157 lakes), and the catchment characteristics that best discriminate between lakes showing recovery (significant increase in alkalinity) and those not showing recovery (no significant increase) were determined. A significant decline in sulphate concentrations occurred in 82–98% of the lakes, depending of the region. Base cation (BC) concentrations decreased for most lakes, but to a lesser extent than those of  $\text{SO}_4$ . Consequently, a significant increase in Gran alkalinity occurred in 40–92% of the lakes. The recovery from acidification has been strongest in lakes in south Finland, where both levels and decrease of S (and N) deposition have been higher compared to other regions. A significant increase in pH was detected in about 50% of the lakes in the south. Here labile aluminium concentrations also decreased in the most acidic lakes. Recovery has occurred most strongly in lakes which have  $\text{SO}_4$  as a dominant acid anion, whereas recovery has been weaker in acidified humic lakes which have organic anion as a dominant acid anion. The non-recovering lakes in south Finland have higher proportion of exposed bedrock in the catchment, and higher TOC and lower BC concentrations. In central Finland the proportion of peatland and TOC concentrations were higher and the decrease of BC concentration was steeper in non-recovering lakes than in recovering lakes. In north Finland, catchment characteristics, trend slopes and concentrations did not separate the recovering and non-recovering lakes. The non-recovering lakes were also located in regions which are acid-sensitive based on bedrock type, soil properties, weathering rate and runoff. These factors have resulted in lower concentrations and steeper downward trends for base cations.

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# 1 Introduction

Acidification of terrestrial and aquatic ecosystems has been among the most serious environmental threats for decades in large areas of Europe and north America. Acidification of rivers and lakes has been a major environmental problem in northern Europe in the Nordic Countries (Finland, Sweden, Norway), where the relative sensitivity of surface waters to acidic deposition is high due to acid-sensitive geology. Sulphur deposition has been the major driving force in the anthropogenic acidification of lakes there (e.g. Skjelkvåle et al., 2001a), resulting in the loss and damage of fish stocks and reduced biodiversity (Tammi et al., 2003a). Due to the implementation of successful emission reduction measures, the total European emissions of SO<sub>2</sub> declined 67% from 1980 to 2000 (Lövblad et al. 2004). As a consequence, S deposition has declined over extensive areas in Europe. In Finland, sulphate deposition has decreased 30% in the north and up to 60% in south Finland since the late 1980s (Vuorenmaa, 2004). Base cation deposition has also declined somewhat. After a decline in the 1980s the base cation deposition leveled out in the 1990s (Ruoho-Airola et al., 2003; Vuorenmaa, 2004; Ukonmaanaho and Starr, 2002). The decline of SO<sub>4</sub> deposition has exceeded the decline of base cation deposition, particularly deposition of Ca and Mg, resulting in the decrease of H<sup>+</sup> and acidifying potential (defined as [SO<sub>4</sub>] – [Ca + Mg]) in deposition throughout the country (Vuorenmaa, 2004).

The decreasing S deposition has resulted in the recovery of sensitive surface waters from acidification (e.g. Stoddard et al., 1999; Evans et al., 2001; Skjelkvåle et al., 2005). In Finland, the regional-scale assessment for the period 1990–1999 showed a statistically significant decline in sulphate concentrations in 80% of the lakes in south and central Finland, and in 60% of lakes in north Finland. A significant increase in Gran alkalinity was detected in 60% of the lakes in the south, but only 20–25% of the lakes in central and north Finland (Forsius et al., 2003). Many lakes in these regions have been exposed to modest load of sulphur deposition and were, therefore, expected to show lower recovery (alkalinity) responses compared to most affected areas in south Finland.

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The site-specific analysis between the deposition and lake data showed that in many lakes changes in alkalinity did not correspond consistently to changes in concentrations of sulphate and charge-balance ANC in bulk precipitation (Forsius et al., 2003), and the variation in alkalinity trends between the lakes within the regions cannot be explained by deposition pattern alone.

Recovery of lakes from acidification can be expected to show different paths, which are partly dependent on catchment attributes. Characteristic for many forested catchments in Finland is thick acidic organic soil horizons under coniferous forests and a high proportion of peatlands, a source of large amounts of humic matter for lakes. Therefore Finnish lakes are greatly influenced by natural organic acids, particularly in central and north Finland (Kortelainen and Mannio, 1990). In the pH range 4-7 a significant fraction of organic acids can be considered strong and have large influences on pH and alkalinity (e.g. Brakke et al., 1987; Munson and Gherini, 1993). Hydrology-induced inputs of organic acids may reflect directly on alkalinity values and suppress the recovery (Mannio, 2001a).

In this paper, the regional recovery of lakes in Finland from acidification is described. The aims are: (i) to present trends in lake water acidification chemistry in south, central and north Finland for the period 1990–2003 and (ii) to determine the catchment characteristics that best discriminate between lakes showing recovery and those not showing recovery.

## 2 Materials and methods

### 2.1 Dataset

#### 2.1.1 Monitoring lakes

The study comprised 157 lakes of the Finnish network of Regional Monitoring of Lake Acidification (RMLA) (Fig. 1) with data for the 14 year period, 1990–2003. A water

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sample from each lake was taken each year, either from the middle of the lake (1 m depth) or at the outlet, during the autumn thermal overturn phase. For 93 of the lakes there is a full set of data for all 14 years, and for 139 lakes have data for 13 years, and the remainder, at least 10 years of data.

5 Catchment characteristics, including lake and catchment area, areas of peatland, exposed bedrock and agricultural land and hydrological type, were determined from topographic maps (1:20 000). The hydrological type of the lake was classified as: seepage (inflow or no inflow, no outflow), headwater (no inflow, outflow), or drainage lake (upstream lakes above a study lake, outflow). Ditching of forest soils is the most  
10 important direct human disturbance in the study catchments. The occurrence of ditching was estimated from maps. The water retention time of the lakes was estimated from lake bathymetry and runoff data for similar catchments (Mannio, 2001b), as runoff for the RMLA lakes has not been measured. The RMLA lakes are mainly small (median area=10 ha) seepage lakes or headwater lakes (117 lakes with no upstream lakes  
15 and 32 drainage lakes with < 5 % upstream lake area in the catchment). Land cover is mainly forest (122 lakes in catchments with no agriculture and only 7 lakes in catchments with > 3 % area under agriculture). Chemically the lakes are acid sensitive with low base cation concentrations (Mannio and Vuorenmaa, 1995).

### 2.1.2 Chemical analyses

20 The water samples were analyzed in the laboratories of Environment Administration using standard methods. The following variables are dealt with in this paper: pH, Gran alkalinity, and concentrations of base cations (BC=calcium (Ca) + magnesium (Mg) + sodium (Na) + potassium (K)), sulphate ( $\text{SO}_4$ ), chloride (Cl), nitrate ( $\text{NO}_3$ ) and total organic carbon (TOC). Measurements of aluminium fractions were only included in  
25 the routine analysis from 1992. Labile Al ( $\text{Al}_{lab}$ ) was calculated as the difference of total reactive and non-labile Al. Ion concentrations are expressed as  $\mu\text{eq l}^{-1}$ , except pH, TOC ( $\text{mg l}^{-1}$ ) and  $\text{Al}_{lab}$  ( $\mu\text{g l}^{-1}$ ). Calculated acid neutralizing capacity, charge-balance ANC ( $\text{ANC}_{CB}$ ), was defined as the equivalent sum of base cations minus the

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equivalent sum of strong mineral acid anions (BC) - ( $\text{SO}_4 + \text{NO}_3 + \text{Cl}$ ). Non-marine fractions of sulphate ( $\text{xSO}_4$ ) and sum of base cations ( $\text{xBC}$ ) were estimated as the differences between total concentrations and concentrations attributable to marine salts, the latter based on ratios to Cl in seawater. Concentrations of organic anions ( $\text{A}^-$ )

5 were estimated from TOC concentrations and pH using the model presented by Kortelainen (1993a). In this model an empirical measure for organic anions has been developed using titration results of isolated hydrophobic and hydrophilic acids (second-order equations as a function of pH) combined with the fractionation results of DOC (proportions of acid fractions). This model describes the dissociation of organic acids, and  
10 requires pH and DOC /TOC concentration of the water samples.

The contribution of strong organic acids to buffering capacity was estimated using the method presented by Lydersen et al. (2004), in which a modified expression for  $\text{ANC}_{\text{CB}}$ , the strong organic acid adjusted ANC ( $\text{ANC}_{\text{OAA}}$ ) in which permanent anionic charge from strong organic acids is included:

$$15 \quad \text{ANC}_{\text{OAA}} = \text{ANC}_{\text{CB}} - (1/3\text{CD}) * \text{TOC}, \quad (1)$$

where CD is charge density of the organic matter ( $\mu\text{eq}/\text{mg}$  of C). A CD value of 9.7, based on studies from Finnish lakes (Kortelainen, 1993a), was used. This value agrees well with the value  $10.2 \mu\text{eq}/\text{mg}$  of DOC) used by Lydersen et al. (2004), reported from the Swedish humic lakes (Hruska et al., 2003).

### 20 2.1.3 Statistical methods

Trends in acidification for the period 1990–2003 were analysed by a regional approach using three geographical regions: south ( $n=61$ ), central ( $n=57$ ) and north Finland ( $n=39$ ) (Fig. 1). The division of the data was based on the monitoring of the long-range transboundary air pollution impacts in lakes reflecting both different environmental conditions and gradients in initial ionic concentrations, deposition levels and trend slopes, decreasing from south-to-north (Mannio and Vuorenmaa, 1995). Monotonic trends for  
25 each study lake were tested using the non-parametric Kendall- $\tau$  test, and a p value of

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<0.05 was used to indicate statistically significant trends. The gradients of the trends (change per year) were calculated using the Sen slope estimation method (Sen, 1968). As the data of  $Al_{lab}$  were limited, they were excluded from the statistical trend analysis. At sites where pH was >6.2 the laboratories did not analyze aluminium. At many sites inorganic bound aluminium was below the detection limit ( $<10 \mu g\ l^{-1}$ ).

The trend of Gran alkalinity was used as a measure of acidification recovery. Alkalinity provides a measure of the buffering capacity of waters, and the most important anions in buffering systems of natural waters are primarily bicarbonate (carbonate system) and weak organic acids.

The lakes within the region were divided into two groups: those having a statistically-significant increase ( $p<0.05$ ) in alkalinity (recovering) and those having no significant increase (non-recovering). These two groups (recovering and non-recovering) were tested for statistically-significant differences in terms of catchment characteristics and other chemical parameters using the Wilcoxon rank-sum (Mann-Whitney) test and discriminant analysis using logistic regression. The latter is frequently used alternative non-parametric application of “classical” discriminant analysis (Press and Wilson, 1978; Tabachnick and Fidell, 1996). The Mann-Whitney test is non-parametric test to compare two unpaired groups, and a p value of <0.05 was used to indicate statistical difference in the population medians. A logistic regression model was applied with ‘recovering’ and “non-recovering” as the dependent dichotomy variable (classes), and the following as independent variables: catchment characteristics lake area (ha), catchment area (ha), catchment-to-lake ratio, proportion of peatland (%), proportion of exposed bedrock area (%), retention time (year), and chemical parameters trend slope values (1990–2003) and concentrations (median 1990–2003) of  $xSO_4$ ,  $xBC$  and TOC. A forward selection procedure was used with a significance level of 0.15 for entry into the model (PROC LOGISTIC, SAS Institute Inc., 1989). In most applications, all variables considered had some discriminatory power. To choose the model that provides the best discrimination using the sample estimates, a moderate significance level is appropriate (SAS Institute Inc., 1989). The Wald Chi-Square test (maximum

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likelihood estimators) was used to test the significance of individual logistic regression coefficients for each independent variable. Measures of the effectiveness (predictive ability) of the model were interpreted by percentages of concordant and discordant pairs. The lakes were paired between the recovery groups and the pair was concordant (discordant) if the lake which belong to the recovery group had a higher (lower) fitted probability belong to the recovery group; ties (no concordant/discordant pairs) occurred when two lakes in the lake pair had the same predictor sets.

### 3 Results

#### 3.1 Regional characteristics of water chemistry

Concentrations of sulphate and base cations in lakes are greatest in south Finland (Fig. 2). In north Finland, the lakes are more dilute with low ion concentrations, and lower  $ANC_{CB}$  and Gran alkalinity.  $ANC_{CB}$  is somewhat higher in central Finland due to higher base cations to sulphate ratio. The concentrations of  $Al_{lab}$  are somewhat higher in south Finland compared to other regions. Based on the median values in 1990–2003, the percentiles (25, median, 75 and 90%) in aluminium concentrations of the lakes were: <10, 10, 40 and  $70 \mu g l^{-1}$  in south, <10, 10, 20 and  $20 \mu g l^{-1}$  in central, and <10, <10, 10 and  $20 \mu g l^{-1}$  in north Finland, respectively. Nitrate concentrations were low in comparison to lakes in the other Nordic countries and central Europe (Skjelkvåle et al., 2001b). 90% of the lakes in all regions had median concentrations  $<2 \mu eq l^{-1}$ . Nitrate concentrations were slightly higher in south Finland (median  $0.8 \mu eq l^{-1}$ ) compared to central and north Finland ( $0.6$  and  $0.4 \mu eq l^{-1}$ , respectively). TOC concentrations were the highest in the lakes of central Finland, where the proportion of peatlands in the catchments is the highest. The median proportion of peatland coverage is 24% in central Finland compared to 7% and 22% in south and north Finland, respectively. Ditching and draining of peatlands have been widely carried out in Finland. The highest proportion of study catchments subjected to ditch-

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ing were in central Finland, 65% of the catchments compared to 16% and 26% of the catchments in south and north Finland, respectively.

## 4 Trends in concentrations 1990–2003

### 4.1 Sulphate and base cations

5 A statistically-significant decreasing trend in  $\text{xSO}_4$  concentrations was detected in 98% of lakes in south Finland, 95% of lakes in central Finland and 82 % of lakes in north Finland (Fig. 3). The slopes of the trend for sulphate were the greatest for lakes in south Finland. The median slope was  $-4.1 \mu\text{eq l}^{-1} \text{yr}^{-1}$ , compared to  $-1.8 \mu\text{eq l}^{-1} \text{yr}^{-1}$  for central Finland and  $-1.4 \mu\text{eq l}^{-1} \text{yr}^{-1}$  for north Finland. The trends in base cation  
10 concentrations were decreasing for most lakes, but to a lesser extent than those of sulphate (Fig. 4). For those lakes with downward trends of  $\text{xBC}$  ( $n=121$ ), in only five lakes in central and in 10 lakes in north Finland  $\Delta\text{xBC} > \Delta\text{xSO}_4$ . A statistically significant decreasing trend for  $\text{xBC}$  concentrations was detected in 23–43% of the lakes, and some lakes exhibited significant increases, depending on the region (Figs. 3,4). The median  
15 annual change for  $\text{xBC}$  concentration was  $-0.97 \mu\text{eq l}^{-1} \text{yr}^{-1}$  in south,  $-0.63 \mu\text{eq l}^{-1} \text{yr}^{-1}$  in central and  $-0.74 \mu\text{eq l}^{-1} \text{yr}^{-1}$  in north Finland.

### 4.2 Gran alkalinity and charge-balance ANC

Gran alkalinity (measured) and  $\text{ANC}_{\text{CB}}$  (calculated) increased significantly in 92% and 82% of the lakes in south, 40% and 42% in central and 49% and 21% in north Finland, respectively (Fig. 3). In the whole study lake set in south Finland, all the slopes of alkalinity and  $\text{ANC}_{\text{CB}}$  in 1990–2003 were increasing, and 84% and 88% of the lakes in central Finland and 97% and 82% of lakes in north Finland showed increasing trends in alkalinity and  $\text{ANC}_{\text{CB}}$ , respectively. The median annual changes for Gran alkalinity  
20 and  $\text{ANC}_{\text{CB}}$  were  $1.9$  and  $2.4 \mu\text{eq l}^{-1} \text{yr}^{-1}$  in south,  $1.0$  and  $1.1 \mu\text{eq l}^{-1} \text{yr}^{-1}$  in central

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and 1.1 and 0.7  $\mu\text{eq l}^{-1} \text{ yr}^{-1}$  in north Finland, respectively.

#### 4.3 pH and aluminium

pH increased significantly ( $\text{H}^+$  decreased) in about half of lakes in south Finland (Fig. 3), and in 30% of all the lakes. The median annual change for  $\text{H}^+$  was  $-0.045 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in south,  $-0.033 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in central and  $-0.013 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in north Finland, respectively. Low pH is associated with high concentrations of labile aluminium. Decreases in acidity were reflected in decreasing labile aluminium concentrations in the formerly most acidic but now recovering lakes (Fig. 5). The decrease was substantial in the lakes in south Finland in which high  $\text{Al}_{\text{lab}}$  concentrations  $>200 \mu\text{g l}^{-1}$  occurred in the early 1990s. The median concentration of  $\text{Al}_{\text{lab}}$  in the study lakes decreased from 40  $\mu\text{g l}^{-1}$  to 10  $\mu\text{g l}^{-1}$  between 1992 and 2003.

#### 4.4 Nitrate

Nitrate concentrations are very much dependent on temperature-driven biological activity, and therefore sensitive to annual and lake-to-lake variation. Nitrate concentrations decreased significantly in 20–60% of the lakes (Fig. 3), and in 70–80% of the lakes decreasing trends were observed, depending on the region. The median annual change for  $\text{NO}_3$  was  $-0.048 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in south,  $-0.024 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in central and  $-0.020 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in north Finland, respectively.

#### 4.5 Total organic carbon (TOC)

Increasing trend slopes for TOC and organic anions ( $\text{A}^-$ ) were found in 85% of the lakes in south Finland, and were significant in 21% and 28% of the lakes, respectively (Fig. 3). The median annual changes for TOC and organic anions were 0.04  $\text{mg l}^{-1} \text{ yr}^{-1}$  and 0.51  $\mu\text{eq l}^{-1} \text{ yr}^{-1}$  in south, 0.0  $\text{mg l}^{-1} \text{ yr}^{-1}$  and  $-0.16 \mu\text{eq l}^{-1} \text{ yr}^{-1}$  in central and 0.02  $\text{mg l}^{-1} \text{ yr}^{-1}$  and 0.15  $\mu\text{eq l}^{-1} \text{ yr}^{-1}$  in north Finland, respectively.

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5 Recovery and catchment characteristics

The combination of catchment characteristics in south Finland that best reveal the differences between two recovery groups were proportions of exposed bedrock, peatland, and catchment-to-lake ratio (Table 2). The non-recovering lake group was characterized by catchments having a higher proportions of exposed bedrock and larger catchment-to-lake ratio (Table 1). The percentile distributions showed, however, only a little difference in proportion of peatland between the groups (Table 1). Concentrations of TOC and base cations distinguished between recovery groups, and the non-recovering lakes exhibited higher TOC and lower xBC concentrations. TOC and exposed bedrock had the highest effect among the independent chemical and catchment variables, respectively. The percent concordant pairs described predicted probability that classification of lakes is successful based on selected independent variables, and they ranged from 84% to 86%, and percent tied pairs were 0.3% (Table 2). Although not selected into the model, the non-recovering lakes have also smaller lake and catchment size and shorter water retention time (Table 1).

Correlation between trends of alkalinity and xSO<sub>4</sub> concentrations in lakes was weaker in central Finland than in south Finland. There is a geographical pattern between the recovery groups in the region of central Finland. The recovering lakes are located mainly in the southern part of the central Finland region, whereas most of the non-recovering lakes are clustered in the northern part (Fig. 1). The catchment characteristics that distinguished between the recovering and non-recovering lakes were: proportional area of peatlands, catchment-to-lake ratio and water retention time (Table 2). Percentage of peatland and catchment-to-lake ratio were higher and retention time was shorter in the non-recovering lake group (Table 1). Concentrations of TOC ( and organic anion) and sulphate distinguished between recovery groups, and the non-recovering lakes exhibited higher TOC (and A<sup>-</sup>) and lower xSO<sub>4</sub> concentrations. For the trend slopes, discriminatory effect was associated with base cations and sulphate, with more downward trends and steeper slopes for xBC, and gentler decreases

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for  $\text{xSO}_4$  in the non-recovering lakes. Mann-Whitney tests were significant for proportion of peatland, catchment-to-lake ratio and retention time and concentrations ( $\text{TOC}$ ,  $\text{xSO}_4$ ) and change in  $\text{xBC}$  concentrations (Table 3). Non-recovering lakes were more acidic than the recovering lakes, indicated by somewhat lower median value of pH and alkalinity, and acid-sensitive determined by lower  $\text{xBC}$  and  $\text{ANC}_{\text{CB}}$  (Table 1). Except for pH, statistical differences were not found for these parameters between the lake groups.

About half of the monitoring lakes in the north Finland did not show significant increase in alkalinity, and these lakes are found in scattered areas in the region (Fig. 1, Fig. 3). The catchment characteristics and magnitude of trends (slopes) did not distinguish between recovering and non-recovering lakes (Table 2). Nonetheless, in concentrations some statistical differences although weak ( $p < 0.1$ ) were found for  $\text{xSO}_4$ , being higher in non-recovering lake group (Table 1, Table 3). These lakes appear to have somewhat lower  $\text{TOC}$  (and  $\text{A}^-$ ) concentrations and higher  $\text{xSO}_4/\text{A}^-$  – and  $\text{xSO}_4/\text{xBC}$ -ratios, having also slightly steeper  $\text{xBC}$  decrease, gentler  $\text{ANC}_{\text{CB}}$  increase, and mostly lower concentrations of  $\text{ANC}_{\text{CB}}$  compared to the recovering lake group.

## 6 Discussion

### 6.1 Regional characteristics and trends in water chemistry

Concentrations of major ions in lakes decrease from south Finland towards the north (Fig. 2). Finland is situated in northern Europe and spans ten degrees of latitude (from  $60^\circ \text{N}$  to  $70^\circ \text{N}$ ) resulting in wide environmental gradients, particularly in atmospheric deposition. Deposition of sulphur from domestic emission sources and of transboundary air pollution have been highest in south Finland, resulting in the gradient in lake  $\text{xSO}_4$  concentrations. Bulk deposition of sulphate in south Finland has been on average 1.4-fold higher compared to central and 2.3-fold higher compared to north Finland (Vuorenmaa, 2004). Base cations ( $\text{xBC}$ ) concentrations in lakes are also high-

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est in south Finland due to higher atmospheric deposition (Ruoho-Airola et al., 2003; Vuorenmaa, 2004) and more intense weathering and ion-exchange from the thicker and better buffered soils (Kämäri et al., 1991). Higher sulphur deposition there has also increased leaching of base cations in runoff (e.g. Kortelainen et al., 1989). In north Finland, the low ion concentrations in lakes are due to both modest load of air pollution and geochemical inactive bedrock overlain by thin layers of coarse-grained till deposits. Inherently acid-sensitive catchments with low buffering capacity are characteristic in many parts of Finnish Lapland (Kähkönen, 1996). Although the proportion of peatlands in the catchment area is also high in north Finland, the TOC concentrations are low compared with central region. The colder climate and consequently longer soil frost period, lower primary production and decomposition, coupled with thinner soil and peat deposits, are likely to decrease the leaching of organic carbon, resulting in lower TOC concentrations in the lakes (Kortelainen, 1993b).

The decline of  $xSO_4$  concentrations is strongest in south Finland, which is due to the combined effect of greatest decline of sulphur both in bulk deposition and dry deposition (Ukonmaanaho et al., 1998; Ukonmaanaho and Starr, 2002) and relatively high initial concentrations in lakes. The decrease of base cation concentrations has also been the steepest in south Finland. The supply of base cations to the lake is reduced due to the steep decrease of mobile sulphate anion. As with sulphate deposition, the base cation deposition has decreased most strongly in south Finland (Forsius et al., 2003; Vuorenmaa, 2004). According to Mannio (2001b), the slope of  $xBC$  trend in lakes is steeper in south Finland than in other regions due to lower proportion of peatland and higher proportion of exposed bedrock in the catchment and longer water retention time (Table 1). These characteristics may have influenced the biogeochemical processes affecting sulphate as well.

The lakes in south Finland have experienced the strongest increase in alkalinity and  $ANC_{CB}$  compared to central and north Finland. A more gentle decline in lake water  $xBC$  concentrations compared to that of  $xSO_4$  (Fig. 4) is a key condition for improving acid-base status and buffering capacity in soil and water. Correspondingly, chemical recov-

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ery of the lakes has been most evident in the southern portions of Sweden and Norway (Skjelkvåle et al., 2005). The non-marine sulphate to organic anion ratio ( $x\text{SO}_4/\text{A}^-$ ) was greater than 1 in 92% of the lakes in south Finland (Fig. 1, Table 1), due to greater sulphur deposition and lower amount of peatlands in the catchments. Decreased sulphur deposition (and acidifying potential in deposition) have caused increases in alkalinity in lakes. Sulphate concentrations exceeded organic anion concentrations in only 30% of the lakes in central and north Finland, and the influence of mineral acids is significantly superimposed on organic acid contributions to acidity in these regions. The signs of increasing alkalinity in north Finland are not reflected in the same way in calculated  $\text{ANC}_{\text{CB}}$ . The small headwater or seepage lakes in north Finland have low ionic strength (with  $x\text{BC}$  typically  $<100\text{ }\mu\text{eq l}^{-1}$ ) that have had only modest impacts from air pollutants. Variation in hydrological regimes can cause pronounced inter-annual variations in ion concentrations, causing 'noise' in time series (Skjelkvåle et al., 2006). Recovery from acidification in the Finnish lakes has continued after 2000. A significant increasing trend in alkalinity occurred in 27% of the monitoring lakes during 1990–1999 (Forsius et al., 2003), whereas alkalinity had significantly increased in 60% of the lakes by 2003. The trend for up to 2003 in the present study showed that the decrease in  $x\text{BC}$  concentrations had slowed down, which confirms the continued positive trends in  $\text{ANC}_{\text{CB}}$  and alkalinity. Nearly all the slopes of alkalinity and  $\text{ANC}_{\text{CB}}$  in 1990–2003 were increasing, which may indicate that regional-scale recovery might be a more common phenomenon than shown by the statistically significant results alone.

The increase of pH was less common than changes in alkalinity. A significant increase in pH (and decrease in  $\text{H}^+$ ) has been most common in south Finland. A significant increase in lake water pH has also been observed in southern parts of Sweden and Norway, and results from individual sites there have shown decreases of labile aluminium concentrations simultaneously with increasing pH (Skjelkvåle et al., 2005). Highest labile aluminium concentrations were found in south Finland, showing also decline with increasing pH.

Earlier studies have suggested that nitrate plays only a minor role in the acidity status

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in Finnish lakes (Mannio, 2001a; Forsius et al., 2003). The nitrate pattern with predominantly decreasing concentrations suggests that there are presently no indications of elevated nitrate levels in forested Finnish lakes. Seasonally monitored lakes in Finland show no or decreasing trends in NO<sub>3</sub> concentrations (Forsius et al., 2001; Kleemola and Forsius, 2006). European nitrogen emissions and nitrogen deposition have been decreasing in Finland during the 1990s (Ruoho-Airola et al., 2004; Vuorenmaa 2004).

Concentrations of TOC has increased in many lakes in south Finland. Increased DOC concentrations over the last two decades have been documented across substantial parts of northern and central Europe (Skjelkvåle et al., 2001a; Evans et al., 2005; Skjelkvåle et al., 2005) and eastern north America (Stoddard et al., 2003). Vuorenmaa et al. (2006) studied changes of TOC concentrations over the period 1987–2003 in 13 seasonally monitored acid-sensitive lakes located throughout the Finland, and ten of the lakes showed a highly significant increase. This may indicate that significant increasing TOC trends might be a more common phenomenon than detected in the present study by only autumn samples alone. A gradual decline of the sulphur deposition is one of the proposed drivers underlying the increased DOC concentrations (Stoddard et al., 2003; Evans et al., 2005; Vuorenmaa et al., 2006). This hypothesis is based on the proposed inverse relationship between mineral acidity and organic acidity (Krug and Frink, 1983), or decreased ionic strength of soil solutions (Tipping and Hurley, 1988; Evans Jr et al., 1988; Vance and David, 1989), which would increase DOC flux. Vuorenmaa et al. (2006) showed that larger initial xSO<sub>4</sub> concentrations, larger decline in xSO<sub>4</sub> concentrations and larger increases in charge-balance ANC have been accompanied by larger increases in TOC. These attributes are common for lakes in south Finland. If increased DOC is primarily related to decreased sulphur deposition, the DOC trends may indicate recovering soil conditions from mineral acid deposition. On the other hand, increase in pH and alkalinity of the most acidified surface waters may be buffered by increasing organic acidity. Several interacting factors may determine the long-term changes in organic carbon in surface waters, but detailed analysis of the drivers is beyond the scope of this paper.

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# 7 Regional recovery patterns and catchment characteristics

## 7.1 South Finland

The non-recovering lakes have a low pH, alkalinity and slightly elevated labile aluminium concentrations (Table 1), and a high proportion of exposed bedrock in the catchment and low lake water xBC concentrations, characteristics for acid-sensitive catchment with poor base cations pool and low neutralization properties. The bedrock and soil in acid-sensitive areas in south Finland are composed of granite or other acidic siliceous rocks, and overburden usually consists of thin soil layers and coarse soil types such as till, sand and gravel (Kämäri, 1986; Nuotio et al., 1990; Huttunen et al., 1990).

The non-recovering lakes have slightly steeper downward trends in xBC and somewhat less steep decrease in  $xSO_4$ , and exhibited smaller increases in  $ANC_{CB}$  than recovering lakes (Table 1). However, the median  $ANC_{CB}$  value of the non-recovering lakes was higher than that of the recovering lake group and trend slopes are increasing. A significant increasing trend in  $ANC_{CB}$  was detected in 40% of the lakes. It is apparent that also other factors than acid-sensitive characteristics in the catchment may have buffered the alkalinity increase. Peatland was important in distinguishing the non-recovering lakes, as indicated by high concentrations of TOC (Table 1, Table 2, Fig. 6). The peatland even in catchments with low peatland coverage are mainly located in the riparian zone of the lakes in the lower part of the catchment, where it contributes to the runoff and lake water chemistry (Schiff et al., 1998). Organic acidity derived from the peatlands is an important contributor to buffering capacity and acidity in non-recovering lakes, as indicated by lower  $xSO_4$  to  $A^-$  and higher  $A^-$  to xBC ratios. Taking the organic anion into account in the sensitivity concept ( $xBC - A^-$ ) (Forsius, 1989), the high content of humic matter increase the acid-sensitivity in these lakes. The level of 'original'  $ANC_{CB}$ , which behaves conservatively to TOC changes (e.g. Neal et al., 1999), is higher but the organic acid adjusted  $ANC_{OAA}$  is lower in non-recovering lakes than in recovering lakes (Table 1, Fig. 6). The inter-annual variation in TOC was reflected in pH and alkalinity values in non-recovering lakes (Fig. 6). The elevated TOC (and  $A^-$ ) con-

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centrations in the early 1990s and in 1997-2001, induced by high autumn runoff events coincided with decrease of pH and depressions of  $ANC_{OAA}$  and alkalinity. There is also one climate-driven possibility, in which ions may be diluted due to elevated water input directly from precipitation and by discharge from the catchment (Clair, 1992; Mitchell et al., 2001).

## 7.2 Central Finland

The lakes in the southern part of central Finland region have received greater sulphate deposition and are also subjected to higher decline of S deposition than the lakes in the northern part, and exhibited both higher  $xSO_4$  concentrations and steeper decline in concentrations in lakes. Sulphate derived minerogenic acidity has been the dominant acidifying agent in these lakes, and decreased mineral acid input has caused more directly increase of buffering capacity in lakes. The non-recovering lakes which are clustered mostly in the northern part, have higher TOC concentrations and higher contribution of organic acids to the acid-base status and acidity, and were somewhat more acidic compared to recovering lakes, indicated by lower median pH values and alkalinity (Table 1, Fig. 6). The non-recovering lakes have lower  $xSO_4$  to  $A^-$  and higher  $A^-$  to  $xBC$  ratios and lower  $(xBC - A^-)$  (Table 1). Subtracting strong organic acid adjusted ANC ( $ANC_{OAA}$ ) from original charge-balance ANC ( $ANC_{CB}$ ), the difference is larger in non-recovering lakes ( $ANC_{CB} - ANC_{OAA}$  median difference  $39 \mu eq l^{-1}$ ) than in recovering lakes (median difference  $23 \mu eq l^{-1}$ ). Kortelainen (1993a) has shown that Gran alkalinity in Finnish lakes is lowered by  $5.3 \mu eq l^{-1}$  for each  $mg l^{-1}$  of TOC, and the average acid-neutralizing capacity provided by organic anion in Gran titrations was estimated to be  $1.6 \mu eq l^{-1}$  for each  $mg l^{-1}$  of TOC. Each  $mg l^{-1}$  of TOC provides thus  $5.3 \mu eq l^{-1}$  of strong and  $1.6 \mu eq l^{-1}$  of weak organic acid anions. Catchment-to-lake ratio was selected as independent to the model, probably due to intercorrelation with proportion of peatland and TOC concentrations (Kortelainen, 1993b).

$ANC_{OAA}$  was better correlated with Gran alkalinity than  $ANC_{CB}$ , particularly in the

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non-recovering lakes (Table 4, Fig. 6). The declines in alkalinity and pH in the early 1990s and in 1998 are probably the result of hydrology-induced organic acid surges.

Ditching had taken place in 82% of the catchments in the non-recovering lake group (39% in recovering lakes), and in most of the catchments ditching took place before 1990. Both runoff and leaching of TOC usually show short-term increase after ditching and level off during the following years (e.g. Kortelainen and Saukkonen, 1998). The role of ditching on organic acid surges and alkalinity patterns was not studied here. In general, the ditching may intensify the water flow to lakes during heavy rainfalls.

While episodes of organic acid surges largely determine the short-term fluctuations in alkalinity, the pool of base cations in soil and trend in base cations are essential in determining the long-term development in alkalinity. The xBC concentrations in most of the recovering lakes increased slightly, suggesting that these soils have not been subjected to strong acidification. In addition, the decline of xBC deposition levelled off during the 1990s, which may have contributed to positive xBC pattern in these lakes. A significant increase in  $ANC_{CB}$  was detected in 80% of the recovering lakes. In contrast, concentrations of xBC in non-recovering lakes have decreased over the study period along with  $xSO_4$ , and has resulted in a little change in  $ANC_{CB}$ . A significant increase in  $ANC_{CB}$  was found in 20% of the lakes in non-recovery group. Base cation deposition decreases towards the northern part Finland region (Vuorenmaa, 2004), but the decrease of BC deposition becomes gentler moving from south towards the northern part (Ruoho-Airola et al., 2003). It is expected that base cation concentrations decrease with sulphate, but several catchment characteristics may have caused xBC concentrations to not increase in the same way as in recovering lake group. The location of the areas of non-recovering lakes agreed well with the areas identified as acid-sensitive based on geochemical properties of bedrock and soil, runoff and relief (Kämäri, 1986; Lahermo et al., 1996), weathering rate from divalent cations  $[Ca+Mg]$  (Johansson and Tarvainen, 1997) and contribution of organic anions  $[xBC-A^-]$  (Forsius, 1989). Finér et al. (2004) have also shown that catchments with high cover of peatlands and usually with flat topography and infertile soils, have lower concentrations and export of base

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cations than in catchments with dominantly mineral soils.

The water retention time in non-recovering lakes was low (median < 1 yr), due to lower depth and water volume (Table 1), and due to higher runoff. The mean runoff (1961–1990) in northern part of central Finland region varied between 350–400 mm yr<sup>-1</sup> whereas in southern part runoff varied between 250–300 mm yr<sup>-1</sup> (Hyvärinen et al., 1995). Longer reaction time between runoff water and soil and longer water renewal time in lakes are known to favour recovery from acidification (Baker and Brezonik, 1988; Nuotio et al., 1990; Schindler et al., 1996), whereas conditions of high discharge and short retention time in the catchment promote dilution of runoff water. The water chemistry in shallow scour lakes draining large, boggy watersheds can be largely affected by runoff induced dilution effect (Clair, 1992). Moreover, processes of in-lake alkalinity generation in short residence time lakes are relatively less important (Baker and Brezonik, 1988).

### 7.3 North Finland

Despite lower S deposition in north Finland, many lakes in this region were subjected to clear anthropogenic acidification during the 1980s. Recovery is evident in lakes in northern Finland (Tammi et al., 2003b; Vuorenmaa et al., 2005) and elsewhere in the Euro-Arctic Barents region due to decreased S deposition (Skjelvåle et al., 2006). Geochemical properties, cold climate and precipitation amounts influence the alkalinity and base cation concentrations and trends in these lakes due to weaker weathering rate of minerals and dilution by precipitation. The leaching of organic acidity is also important in such poorly buffered lakes (Kähkönen, 1996). Vulnerability to acidification therefore varies greatly, even among neighbouring catchments resulting in spatially mosaic-like recovery map. The non-recovering lake group was somewhat more acidic compared to recovering lakes, as indicated by lower median pH values and alkalinity (Table 1, Fig. 6).

Autumn 1992 and 1998 were very wet periods also in north Finland, resulting in high autumn runoff there. High TOC concentrations and low alkalinity, pH and ANC<sub>OAA</sub> in

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the non-recovering lakes were therefore observed in these two years (Fig. 6). While alkalinity tended to increase (19 out of the 20 lakes with increasing slope) in the non-recovering lake group, confounding factors such as elevated runoff and peaks in organic acidity have suppressed alkalinity in most of the acid-sensitive lakes. Proportions of peatlands were estimated to be similar between the lake groups, but  $\text{xSO}_4$  concentrations and  $\text{xSO}_4/\text{A}^-$ -ratios were higher in the non-recovering lake group. The estimate of peatland area is based on topographic maps, and may not give an accurate measure of the total effective amount of organic soils in the catchment. Mineral soils with lower sulphate retention ability and probably lower BC supplies may therefore be more important in the non-recovering lakes than expected on the basis of peatland area estimates.

The long-term monitoring data presented here provide clear evidence that reductions in sulphur emissions and deposition during the last two decades have resulted in a large-scale improvement in water quality and that chemical recovery is proceeding in Finnish lakes. The recovery from acidification has been strongest in lakes in which sulphate has been the major acidifying agent. These lakes are located mostly in south Finland. Many catchments in Finland, particularly in central Finland, have a high proportion of peatlands and high TOC concentrations, and the influence of strong mineral acids is superimposed on organic acid contributions to acidity. Runoff-induced surges of organic acids have suppressed recovery of buffering capacity in many acidified humic lakes. Base cation concentrations are still declining, and this has resulted in lower  $\text{ANC}_{\text{CB}}$  and alkalinity increase in many lakes, particularly in the central and north Finland. Climate change may increase the mineralization of soil organic matter and change the frequency and magnitude of runoff and organic acid episodes. An increase of these types of confounding effects on pH and alkalinity recovery may therefore be anticipated in the future.

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**Table 1.** Percentiles (25, 50 and 75%) for catchment characteristics and for trend slopes and concentrations in the recovering and non-recovering lakes.

Variable	Unit	south Finland (n=61) Recovering (n=56)			Non-recovering (n=5)			central Finland (n=57) Recovering (n=23)			Non-recovering (n=34)			north Finland (n=39) Recovering (n=19)			Non-recovering (n=20)		
		25%	50%	75%	25%	50%	75%	25%	50%	75%	25%	50%	75%	25%	50%	75%	25%	50%	75%
Lake	ha	7	16	36	2	4	29	5	10	43	4	7	15	4	9	24	3	10	27
Catchment	ha	54	92	306	28	62	175	53	111	256	58	113	254	76	115	183	47	112	284
Catchm./Lake		4.6	6.4	8.4	12.5	15.5	16.3	4.3	7.5	13	6.5	13.9	22	6.0	11.3	17	7.2	10.4	18.2
Exp.	%	0.5	5	16	10	29	30	0	0	1	0	0	0	0	0	1	0	0	0
bedrock																			
Peatland	%	4	8	12	4	7	15	4	14	25	23	32	42	8	21	50	3	22	35
Retention	year	1	3	4	0.2	0.9	1.2	0.6	1.6	3.5	0.3	0.8	1.1	0.1	0.2	0.7	0.1	0.3	0.7
time																			
max depth	m	4.3	8.8	13.7	2.5	6.0	12	5.4	8.5	10.1	3.4	5.6	8.6	2.0	2.0	3.0	2	2	4
pH	pHunit yr <sup>-1</sup>	0.004	0.020	0.033	0.000	0.005	0.017	0.000	0.020	0.047	0.000	0.000	0.017	0.000	0.000	0.015	0.000	0.000	0.013
H <sup>+</sup>	μeq l <sup>-1</sup> yr <sup>-1</sup>	-0.20	-0.05	-0.01	-0.19	-0.02	0.00	-0.14	-0.05	0.00	-0.13	-0.02	0.00	-0.03	-0.02	-0.003	-0.14	-0.004	0.000
Alkalinity	μeq l <sup>-1</sup> yr <sup>-1</sup>	1.50	1.95	2.91	1.28	1.50	1.55	1.18	2.00	3.14	-0.33	0.50	1.00	1.12	1.38	1.72	0.62	0.80	0.97
xBC	μeq l <sup>-1</sup> yr <sup>-1</sup>	-1.96	-0.91	-0.39	-2.58	-1.01	-0.97	-1.11	0.14	1.50	-1.57	-1.04	-0.46	-1.30	-0.72	0.00	-1.64	-1.01	-0.37
xSO <sub>4</sub>	μeq l <sup>-1</sup> yr <sup>-1</sup>	-5.56	-4.14	-2.79	-4.52	-3.91	-3.68	-2.61	-2.00	-1.44	-2.16	-1.76	-1.53	-1.68	-1.44	-0.90	-1.97	-1.38	-0.80
ANC	μeq l <sup>-1</sup> yr <sup>-1</sup>	1.73	2.62	4.09	1.48	1.50	2.41	1.10	2.32	4.00	0.15	0.73	1.40	0.52	0.90	1.33	0.00	0.49	0.82
ANC <sub>OMA</sub>	μeq l <sup>-1</sup> yr <sup>-1</sup>	1.77	2.53	3.70	1.48	1.95	2.29	1.13	2.21	3.88	0.29	0.70	1.55	0.36	0.66	1.15	0.01	0.42	1.02
A <sup>-</sup>	μeq l <sup>-1</sup> yr <sup>-1</sup>	0.20	0.51	1.16	-0.44	-0.36	1.50	-0.47	0.36	0.59	-1.04	-0.39	0.35	-0.75	0.35	0.94	-0.56	-0.12	0.34
TOC	mg l <sup>-1</sup> yr <sup>-1</sup>	0.01	0.04	0.10	-0.08	0.01	0.11	-0.05	0.02	0.08	-0.15	-0.03	0.04	-0.09	0.03	0.11	-0.07	-0.01	0.04
pH		5.5	5.9	6.2	5.3	5.6	5.8	5.7	5.9	6.4	5.1	5.5	6.1	5.8	6.0	6.3	5.1	5.8	6.4
Alkalinity	μeq l <sup>-1</sup>	8	27	54	14	21	21	21	36	58	-2	18	49	8	21	32	-3	5	35
xBC	μeq l <sup>-1</sup>	125	170	223	138	161	168	78	167	203	101	126	176	50	64	91	35	58	97
xSO <sub>4</sub>	μeq l <sup>-1</sup>	85	106	134	74	79	99	42	61	79	32	39	49	16	19	34	21	32	38
ANC	μeq l <sup>-1</sup>	32	62	97	67	78	82	32	105	138	52	87	121	31	49	67	7	30	66
ANC <sub>OMA</sub>	μeq l <sup>-1</sup>	13	43	71	28	31	34	22	61	100	19	40	86	8	26	43	-8	8	41
A <sup>-</sup>	μeq l <sup>-1</sup>	29	40	69	83	105	117	24	55	95	58	90	128	31	44	67	20	35	53
TOC	mg l <sup>-1</sup>	3.9	5.3	8.8	11	14.5	15	3.1	7.3	12.2	8.0	12.6	16.2	4.1	5.5	8.1	2.8	4.8	7.7
NO <sub>3</sub> -N	μeq l <sup>-1</sup>	0.43	0.77	1.38	0.54	0.57	1.44	0.36	0.50	0.86	0.43	0.71	1.00	0.36	0.36	0.36	0.36	0.36	0.81
Al lab	μg l <sup>-1</sup>	< 10	10	40	24	25	32	< 10	10	20	< 10	10	20	< 10	< 10	< 10	< 10	10	11
xSO <sub>4</sub> /A <sup>-</sup>		1.46	2.38	3.91	0.58	0.61	1.21	0.44	0.97	2.28	0.34	0.45	0.86	0.33	0.43	0.64	0.36	0.82	1.94
A <sup>-</sup> /xBC		0.19	0.27	0.39	0.50	0.53	0.72	0.21	0.37	0.56	0.50	0.75	0.88	0.51	0.75	0.88	0.38	0.69	0.95
xSO <sub>4</sub> /xBC		0.53	0.63	0.76	0.46	0.47	0.60	0.28	0.37	0.55	0.24	0.34	0.41	0.21	0.30	0.45	0.27	0.48	0.79
(xBC)-(A <sup>-</sup> )	μeq l <sup>-1</sup>	86	113	163	43	87	97	59	71	104	9	26	73	10	22	41	2	19	48

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**Table 2.** Results of logistic regression for catchment characteristics, concentrations and trend slopes. Ordinal number of variable for entry into the model (Number in), Wald Chi-Square test score (Wald Chi-Sq) and associated *p*-value (Pr<Chi-Sq) and percents of concordant and discordant pairs are shown on the model results (n.e.=no entry into the model)

Variable	south				central				north			
	Number in	Wald Chi-Sq	Pr>Chi-Sq	Concordant/Discordant (%)	Number in	WaldChi-Sq	Pr>Chi-Sq	Concordant/Discordant (%)	Number in	Wald Chi-Sq	Pr>Chi-Sq	Concordant/Discordant (%)
Catchment	n.e.			84.3/15.0	n.e.			87.6/12.4	n.e.			
Lake	n.e.				n.e.				n.e.			
Catchment	n.e.				n.e.				n.e.			
Catchment/Lake	3	1.7	0.136		2	1.5	0.218		n.e.			
Exposed	1	4.3	0.039		n.e.				n.e.			
bedrock												
Peatland	2	2.4	0.194		1	10.1	0.002		n.e.			
Retention time	n.e.				3	2.4	0.100		n.e.			
Slope								81.2/18.5				
xBC	n.e.				1	10.6	0.001		n.e.			
xSO <sub>4</sub>	n.e.				2	5.1	0.024		n.e.			
TOC	n.e.				n.e.				n.e.			
Concentration				86.4/12.9				76.2/23.7				66.3/32.1
xBC	2	3.1	0.078		n.e.				n.e.			
xSO <sub>4</sub>	n.e.				1	5.4	0.021		1	3.0	0.083	
TOC	1	6.5	0.011		2	4.1	0.043		n.e.			

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**Table 3.** Comparison of the catchment characteristics, trend slopes and concentrations of recovering and non-recovering lake groups by Wilcoxon rank-sum (Mann-Whitney) test for central and north Finland (south Finland was ignored in the analysis). A statistically significant difference ( $p < 0.05$ ) is highlighted in bold.

Variable	Unit	Region	
		central	north
Lake	ha	0.0742	0.8325
Catchment	ha	0.9352	0.9328
Catchment/Lake		<b>0.0248</b>	0.8441
Exposed bedrock	%	0.3989	0.5802
Peatland	%	<b>0.0001</b>	0.5389
Retention time	Year	<b>0.0036</b>	0.8550
xBC	$\mu\text{eq l}^{-1} \text{ yr}^{-1}$	<b>0.0021</b>	0.1865
xSO <sub>4</sub>	$\mu\text{eq l}^{-1} \text{ yr}^{-1}$	0.3055	0.9664
TOC	$\text{mg l}^{-1} \text{ yr}^{-1}$	0.0774	0.1961
pH		<b>0.0084</b>	0.1359
Alkalinity	$\mu\text{eq l}^{-1}$	0.1302	0.1475
xBC	$\mu\text{eq l}^{-1}$	0.3054	0.7148
xSO <sub>4</sub>	$\mu\text{eq l}^{-1}$	<b>0.0034</b>	0.0676
ANC <sub>CB</sub>	$\mu\text{eq l}^{-1}$	0.8708	0.2732
TOC	$\text{mg l}^{-1}$	<b>0.0311</b>	0.5363
xSO <sub>4</sub> /A <sup>-</sup>		<b>0.0033</b>	0.2323
A <sup>-</sup> /xBC		<b>0.0001</b>	0.6734
xSO <sub>4</sub> /xBC		0.1930	0.0917

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**Table 4.** Correlations (Pearson's correlation coefficients) for Gran alkalinity with concentrations of charge-balance ANC ( $\text{ANC}_{\text{CB}}$ ) and strong organic acid adjusted ANC ( $\text{ANC}_{\text{OAA}}$ ) in the different regions (south, central, north Finland) and lake groups (recovering, non-recovering) of RMLA lakes. A statistically significant correlation is denoted with asterisks (  $p<0.0001^{***}$ ,  $p<0.001^{**}$ ,  $p<0.01^{*}$ ).

Region	Lake group	<i>n</i>		$\text{ANC}_{\text{CB}}$	$\text{ANC}_{\text{OAA}}$
south	Recovery	56	Gran alk.	0.914 <sup>***</sup>	0.974 <sup>***</sup>
	Non-recovery	5	Gran alk.	0.964 <sup>*</sup>	0.996 <sup>**</sup>
central	Recovery	23	Gran alk.	0.841 <sup>***</sup>	0.951 <sup>***</sup>
	Non-recovery	34	Gran alk.	0.773 <sup>***</sup>	0.949 <sup>***</sup>
north	Recovery	19	Gran alk.	0.751 <sup>**</sup>	0.937 <sup>***</sup>
	Non-recovery	20	Gran alk.	0.747 <sup>**</sup>	0.929 <sup>***</sup>

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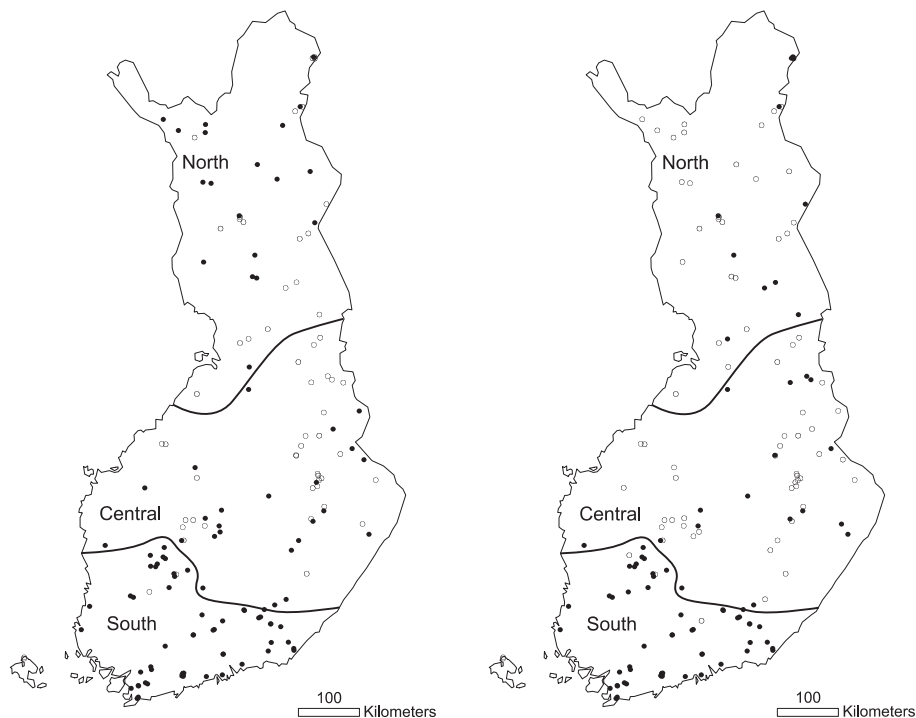
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**Fig. 1.** Location of the RMLA lakes ( $n=157$ ). Location of the three regions (south, central and north) is also shown. Significant increasing trend ( $p<0.05$ ) (•) and no trend (○) in alkalinity in the study lakes 1990–2003 (left map) and non-marine sulphate to organic anion ratio (median value in 1990–2003),  $xSO_4 \geq A^-$  (•) and  $xSO_4 < A^-$  (○) (right map).

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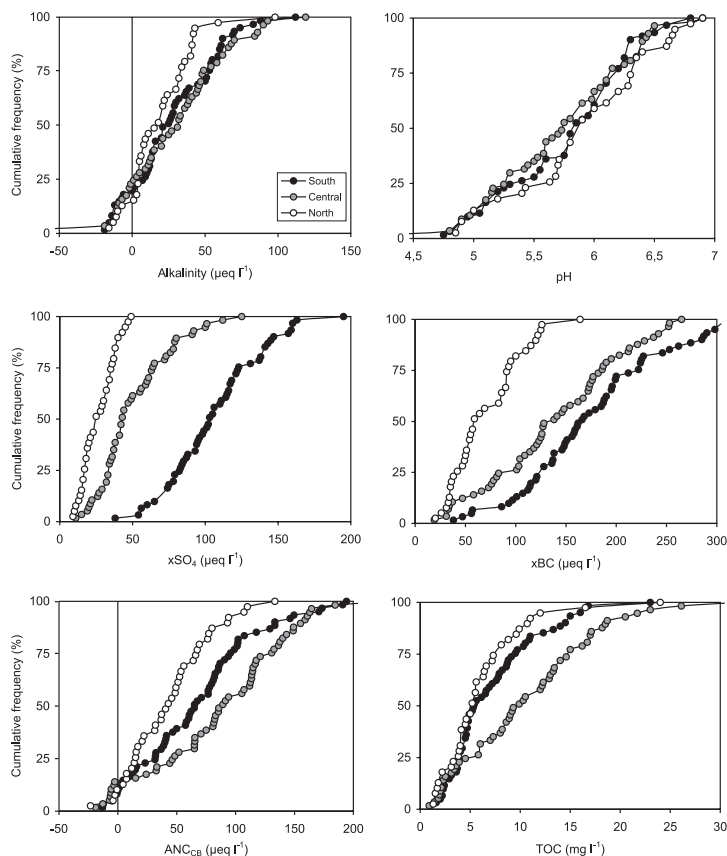
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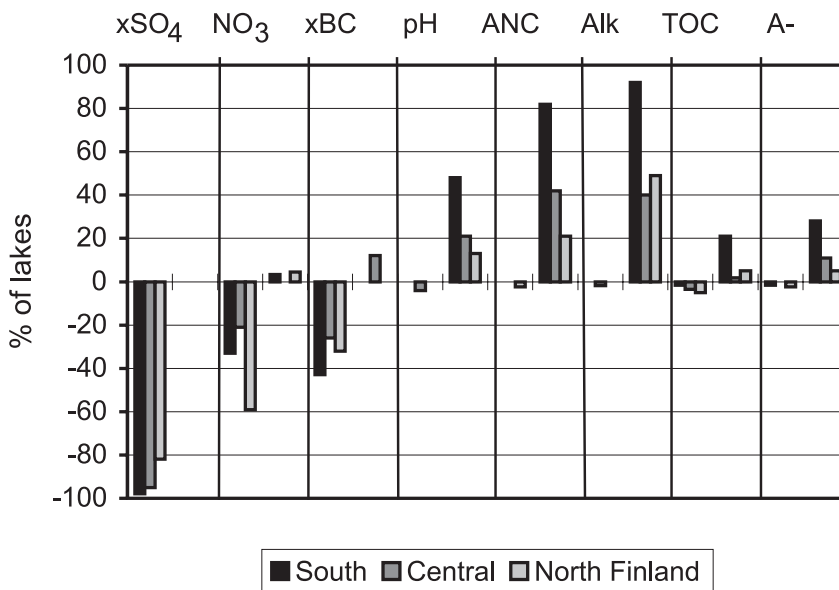


**Fig. 2.** Cumulative frequency distributions (%) for Gran alkalinity, pH,  $xSO_4$ ,  $xBC$ ,  $ANC_{CB}$  and TOC of lakes in south (•), central (◐) and north (◑) Finland. The value for a lake is the median in 1990–2003.



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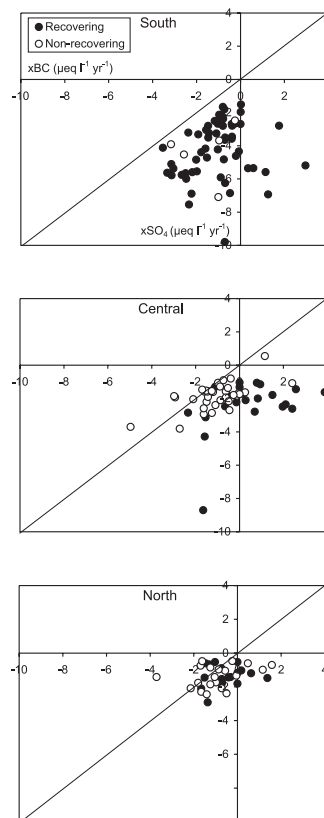


**Fig. 3.** Percent of RMLA lakes (n=157) in south, central and north Finland showing significant increasing or decreasing trends (Kendall-t,  $p < 0.05$ ) of key water quality variables in 1990–2003.

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**Fig. 4.** Relationships between trend slopes (Sen slope estimator, years 1990–2003) for  $xSO_4$  and  $xBC$  in south, central and north Finland. Lakes with significant increase in alkalinity ( $\bullet$  = recovering) or with no trend ( $\circ$  = non-recovering) are separated.

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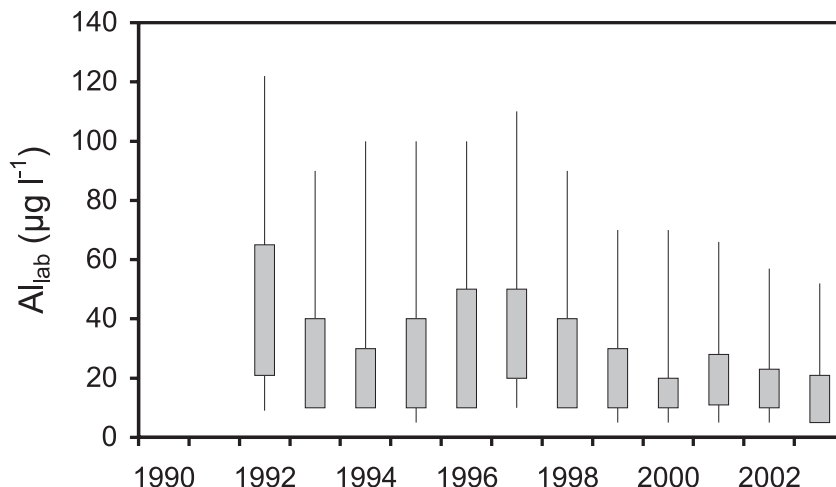
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**Fig. 5.** Trend in labile aluminium concentrations in 65 RMLA lakes in 1992–2003. Lower line = 5th percentile; lower and upper ends of the boxes = 25th and 75th percentiles; upper line=95th percentile. Median value of labile aluminium concentration in 1992 is  $40 \mu\text{g l}^{-1}$  and in 2003  $10 \mu\text{g l}^{-1}$ . The trend data include lakes with median (1992–2003) aluminium concentration  $\geq 10 \mu\text{g l}^{-1}$ .

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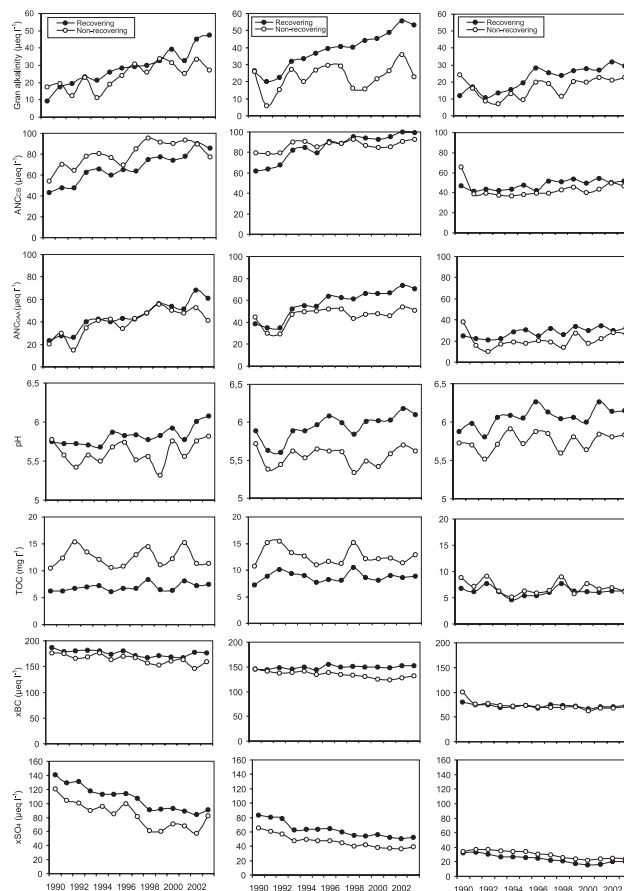
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**Fig. 6.** Time series of annual mean values for Gran alkalinity,  $\text{ANC}_{\text{CB}}$ ,  $\text{ANC}_{\text{OAA}}$ , pH, TOC, xBC and  $\text{xSO}_4$  of recovering lakes (●) and non-recovering lakes (○) in south (left), central (middle) and north (right column) Finland.